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Erwin Pink

(NASA-TT-F-15635) ALLOY SOFTENING IN BODY-CENTERED CUBIC METALS (Linguistic Systems, Inc., Cambridge, Mass.) 41 p HC $5.25 CSCL 11F

N74-27017

G3/17 Unclas 40674

Translation of: "Legierungsentfestigung in kubisch-raumzentrierten Metallen." Legierungsentfestigung(Alloy Softening), Vol. 64, No. 12, 1973, pp. 871-881.

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON, D.C. 20546 JUNE 1974

STANDARD TITLE PAGE

1. Report No.	2. Government Accession No.	3. Recipient's Catalog No.
NASA TT F-15635		

4. Title and Subtitle	5. Report Date
ALLOY SOFTENING IN BODY-CENTERED CUBIC METALS	June, 1974
	6. Performing Organization Code

7. Author(s)	8. Performing Organization Report No.
Erwin Pink	10. Work Unit No.

9. Performing Organization Name and Address	11. Contract or Grant No.
LINGUISTIC SYSTEMS, INC. 116 AUSTIN STREET CAMBRIDGE, MASSACHUSETTS 02139	NASW-2482
	13. Type of Report & Period Covered TRANSLATION
12. Sponsoring Agency Name and Address NATIONAL AERONAUTICS AND SPACE ADMINISTRATION WASHINGTON, D.C. 20546	14. Sponsoring Agency Code

15. Supplementary Notes

Translation of: "Legierungsentfestigung in kubisch-raumzentrierten Metallen." Legierungsentfestigung Vol. 64, No. 12, 1973, pp. 871-881.

16. Abstract

In the range of thermally activated deformation of bcc metals, alloying with substitutional and interstitial elements can cause a reduction of the critical resolved shear stress. In iron alloys this occurs between 100 and 300°K. The theories are manifold. Apparently alloy softening cannot be explained by one single mechanism. Depending on the type of alloy, the easy nucleation of double kinks, scavenging, and the resulting change in deformation mechanism may be responsible.

17. Key Words (Selected by Author(s))	18. Distribution Statement
	UNCLASSIFIED - UNLIMITED

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price
UNCLASSIFIED	UNCLASSIFIED		

ALLOY SOFTENING IN BODY-CENTERED CUBIC METALS

by Erwin Pink

(From the Erich Schmidt Institute for Solid-State Physics
of the Austrian Academy of Sciences in Leoben)

---ABSTRACT---

In the range of thermally activated deformation of bcc metals, /871* alloying with substitutional and interstitial elements can cause a reduction of the critical resolved shear stress. In iron alloys this occurs between 100 and 300°K. The theories are manifold. Apparently alloy softening cannot be explained by one single mechanism. Depending on the type of alloy, the easy nucleation of double kinks, scavenging, and the resulting change in deformation mechanism may be responsible.

Alloy softening (shortened to 'AS' in the following text) is limited to low temperature deformation. Although hardening predominates in the case of face-centered cubic (f. c. c.) metals, unexpected softening phenomena were nevertheless observed in low-percentage b. c. c. alloys. The concept of 'alloy' is to be understood here in its broadest sense; it includes not only materials with substitutionally dissolved elements, but also interstitial solutions and materials with incompatible excretions.

AS has become known principally through studies of the dependence of mechanical properties on temperature. Two methods were used: hardness tests and, for the most part, tensile tests.

In tests in which temperature (T) and stretching speed ($\dot{\varepsilon} = \phi\dot{\gamma}$) influence the properties, thermal activation analysis can be used. Concepts such as area of activation (A*), volume of

*Numbers in the margin indicate pagination in the foreign text.

activation ($v^* = bA^*$), enthalpy of activation (ΔH), or frequency factor (γ_o) can be hypothesized [1-3].

We should now refer to the usual separation of the externally applied tension (for example the yield point $\sigma_s = \tau_s/\mu$) into two shear components τ_G and τ^*. The non-thermal part τ_G originates in the long-range elastic tension fields (around alloys, excretions, inhomogeneities, etc.). The thermally activated part, the 'effective tension' τ^* derives from short-range hindrances, which are effective only a little farther out than the Burger vector.

Alloying affects τ_G as well as τ^*. Without exception, τ_G is increased, as described by various theories. AS manifests itself, on the other hand, in the lowering of the effective tension τ^*[†]) or also of the total tension τ_s. A basic condition for this is the reduction of the temperature dependence $\partial\tau_s/\partial T \approx \partial\tau^*/\partial T$. Curves with continuously diminishing $\partial\tau_s/\partial T$ are known, for example, for the tantalum-niobium [6] series of mixtures with complete solubility. So, we only speak of true AS when an alloying of small amounts of a second metal leads to a decrease of τ_s (or τ_s^*) and $\partial\tau_s/\partial T$, instead of to the expected increase in strength. Only on further additions of the alloy metal does the expected alloy hardening take place.

If τ_s is chosen as the comparison parameter in the display of temperature dependence, the AS effect can be veiled if the increase in τ_G is stronger than the reduction of $\partial\tau_s/\partial T$. In spite of a reduction in τ^* at low temperatures, the τ_s of the alloy can still be greater than τ_s of the basic metal. Therefore,

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[†]This viewpoint is almost exclusively represented in the literature. An additional influence of the alloy density on the non-thermal component of alloys has only once been taken into consideration [4], yet this stands in contrast to many research results [5].

only the magnitude of the thermal component τ^* serves as a criterion for AS. Whenever τ_s is specified, it must be considered along with $\partial\tau_s/\partial T$.

Some Experimental Results

AS was earlier demonstrated in substitution alloys of iron; since then the literature has assumed huge proportions [7]. The following presentation will cite only the most essential results. The alloys of iron were examined especially frequently [5, 8-26]. Other alloy groups encompass the metals of Group Va [4, 27-36] and of Group VIa [37-41] of the periodic system. AS is characteristic of single crystals as well as of polycrystalline materials.

A typical example of the temperature dependence of the yield point of iron-nickel/silicon substitution alloys [21] is shown in Fig. 1. AS is visible below 200°K if the total yield point is referred to. The effect is made even clearer by use of the effective yield point.

A further instructive way of showing AS is a plot of strength vs. concentration (c). The temperature is also specified as a parameter. Fig. 2 shows this in the case of the hardness of molybdenum-rhenium alloys [41].

At low temperatures, the minimum hardness is strongly marked, then it gradually disappears, and is missing entirely at high temperatures. Hardness as a measure of strength contains both components τ_G and τ^*. One could also choose here only the effective (thermal) part for comparison. Then the softening would be even more significant.

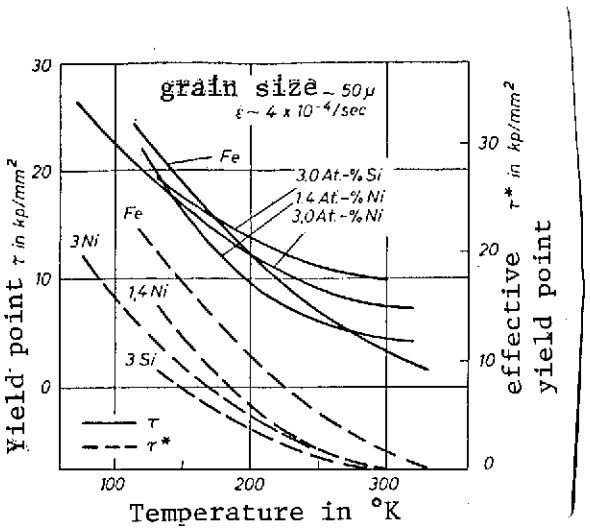

Fig. 1. The temperature dependence of the total and the effective (thermal) shearing stress for polycrystalline iron-nickel/silicon alloys [21].

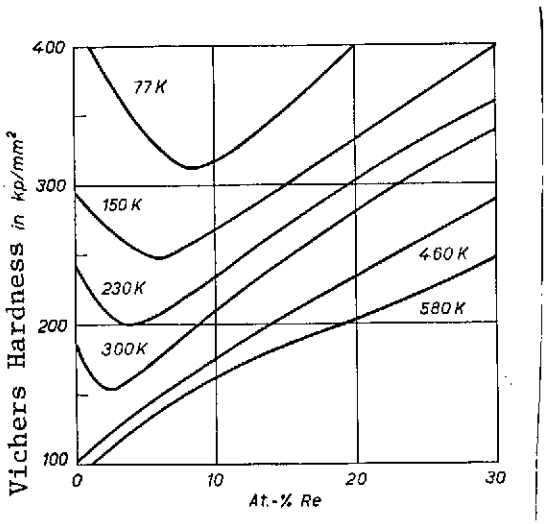

Fig. 2. The concentration dependence of AS in polycrystalline molybdenum-rhenium alloys [41].

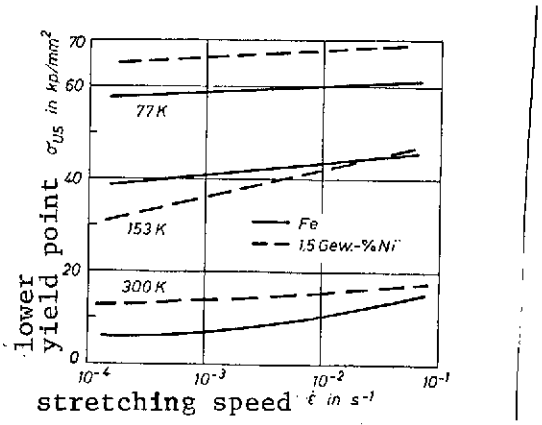

Fig. 3. The dependence of tenacity on the stretching speed in polycrystalline iron-nickel alloys [16].

Fig. 3 shows that thermally activated changes of strength /871
can be noticeable also in σ - log $\dot{\varepsilon}$ - curves [16]. The AS at 150°K shows up not only in that the alloys have lower yield points than pure iron, but also manifest themselves in a greater dependence of the yield points of the alloys on $\dot{\varepsilon}$. As Fig. 3 also shows, both phenomena disappear if the temperature is lowered below 100°K.

AS caused by interstitially dissolved elements was first observed only some ten years ago. The first indications came from tests with decarbonized iron [42, 43], and later came research on iron systematically alloyed with nitrogen [44-46], and on niobium alloyed with oxygen [47]. Fig. 4a shows the results with iron-nitrogen alloys [45]. The room temperature strength climbs continuously with the nitrogen content, but the temperature dependence of the alloys is reduced at medium temperatures, so that AS takes place. As shown in Fig. 4b, the softening disappears below 100°K.

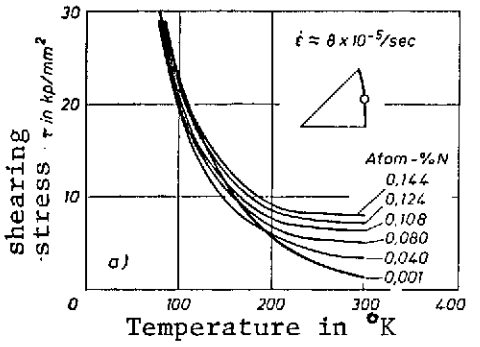

Fig. 4a. AS in iron-nitrogen single crystals [45]. The temperature dependence of the critical shear stress.

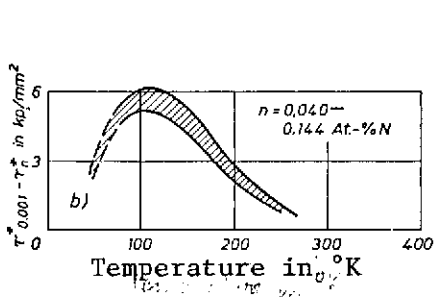

Fig. 4b. AS in iron-nitrogen single crystals [45]. The difference between the effective shear stress of pure iron and of alloys.

A real difference in behavior between substitution- and interstitial mixed crystals is the portion of the alloy that effects a strength minimum in σ_s-c diagrams such as Fig. 2 under otherwise comparable conditions. In interstitial alloys, the minimum is found at about 0.02 to 0.05 Atom-%, while in substitution alloys from 2 to 10 Atom-% is necessary, according to the alloy type and temperature (the point of the minimum is temperature-dependent; see Fig. 2). In one special case, however, when substitutional phosphorus is used, a maximum is found at 0.4 Atom-%. All these data refer to tests with small speeds of deformation (mostly 10^{-5} to 10^{-3}/s).

AS was found especially in dispersion-hardened materials and in metals with 'interstitial' impurities which exceed the solubility limit. Thus, for example, $\partial\sigma_s/\partial T$ is reduced in molybdenum-Al_2O_3 and molybdenum-MgO mixtures [48]. Higher-percentage niobium-oxygen alloys, which probably contain oxide excretions, also show AS.

Theories of Alloy Softening

Theories must primarily keep in mind that AS takes place only in the region of thermally activated alloying activity, therefore below the limiting temperature T_o for maximum thermal activation ($T_o \approx 0.15\ T_s$ at low speeds of stretching). A first simple test for the usefulness of each theory is its applicability to single crystals and polycrystalline materials. Models that take into account only the properties of single crystals (e.g.,of single glide planes) or of polycrystalline materials (e.g., the checking of compatability conditions through activation of a large number of glide systems) [20], are eliminated at the outset.

There have been instances of considering AS in substitutional and interstitial mixed crystals separately from one another [49, 50]. The different effective alloy concentrations and, according to the thinking of several authors, the disappearance of AS under 100°K exclusively in interstitial alloys, were taken as pointing to fundamentally different mechanisms of deformation. The study of substitutional iron-phosphorus alloys [26] has shown, however, that a strength minimum at very small concentrations is not a characteristic limited to interstitial alloys. Furthermore, /873 various workers have shown that AS also disappears under 100°K in several substitution alloys (e.g. Fig. 3) [16, 25, 27, 35]. Because of the similarities of the phenomena in both types of alloys, we should assume for the moment that the origins of AS may be the same. However, since the similarity is not documented

for all substitutional cases (see for example Figs. 1, 2, 10), there exists the possibility that characteristic differences may not derive from the contrast of interstitial to substitutional, but that different substitutional groups exist which differ among themselves in the effects of their admixture in an alloy.

The controversies that persist even today in the area of low temperature deformation reflect very important considerations for AS. They proceed either from modifications of a mechanism of deformation specific ('intrinsic') for b. c. c. crystals, or from the assumption of a mechanism ('extrinsic') influenced by impurities. In the following, the most pertinent works and the theories set forth in them will be described.

The influence of Peierl's Tension or the Double Kink Mechanism. The force for displacement of the dislocation line explained by the periodicity of the lattice leads to Peierl's Tension τ_p. It is proportional to the shear modulus G and the value exp(-w) (w characterizes the extent of the dislocation nucleus) [51]. The change of Peierl's Tension through alloy formation is possible chiefly as a result of these two quantities G and w. The changing of τ_p could only lead to AS if it were non-regular, i.e.,if τ_p decreased only initially with small admixtures of alloying material, but later climbed again in the manner of mixed-crystal hardening. G and w must also therefore change in a non-regular way, i.e., drop initially.

Although according to one theoretical calculation [52] the elastic constant in alloys should be lowered if the modulus of the added alloy metal is higher, nevertheless studies of tantalum-rhenium [28] and niobium-zirconium/vanadium/tantalum alloys [53] have shown that the elastic constants climb readily or follow the tendency towards AS only to an insignificant degree. On the other hand, there is AS even in cases where the

great difference of the moduli might lead us to expect G to be strongly increased (niobium-molybdenum [4, 33, 34], tantalum-tungsten [34]). We are not reckoning with an influence of the elastic quantities in alloy-softened interstitial alloys [54]. Another argument for the influence of the shear modulus G was its influence on the enthalpy of activation (ΔH should be directly proportional to G [14, 31]). The actual reduction of ΔH (for example in the system iron-nickel/silicon [5, 21] or tantalum-niobium [31]) is for the most part greater than would be expected from a possible change in G.

The effect of w is not known for certain. Change in w, for example, local disorders in the neighborhood of the dislocation center that lead to a decrease in τ_p, were imposed on the known lattice expansion of alloys [14].

The direct effect on the Peierl's Tension cannot explain the disappearance of AS at low temperatures. A proof of its presence would be, therefore, the failure of a convergence of the τ - T curves of pure and adulterated (alloyed) metals below 100°K. This is significantly visible in Fig. 1. The actual experimental proof is lacking nevertheless.

The breakdown energy can be reduced by alloy formation, and by reduction of the total energy of the dislocation lines, the cleavage to semi-dislocations can result. The Peierl's potential to overcome the partial dislocations has now become smaller, and the necessary shearing stress is reduced. In fact, the magnitude of the Peierl's Tension is not decisive in the case of a cleavage. Before transverse slip can take place, the cleavage must be made ineffective, and the flow tension will be determined by this process. Detailed theories have been developed for b. c. c. metals [55-57]. We will come back to them later.

At finite temperatures, a thermally activated double kink mechanism can contribute to the dislocation slip [58-59]. In this case, the motion is produced by the formation of a 'double kink,' i.e., a dislocation segment of length $\sim b$ reaching above the Peierl's Tension. The dislocation as a whole proceeds through lateral expansion of a kink into the next Peierl valley. A model has been developed to illustrate the experimentally found deviation of the double kink mechanism in the region of maximum thermal activation, which involves the influence of long-range internal tensions [60]. It is assumed that τ_G varies locally (positively or negatively), and can be given a definite value only on the average. The thermally activated jumping over of short-range barriers will be locally easier or more difficult for a dislocation, which during its wandering through the lattice perceives both the different magnitudes and different directions of the long-range forces, and the speed of the dislocation is thereby influenced. Owing to a mathematical relationship the activation surface A* changes considerably and the activation enthalpy ΔH only slightly; and these activation parameters and the reduced effective tension τ^* comply with the condition

$$\tau^* = \frac{1}{bA^*}\left(\Delta H_o - kT \ln(\dot{\gamma}_o/\dot{\gamma})\right) \qquad (1)$$

Since the effectiveness of the internal tensions decreases at low temperatures according to this theory (only true for $\tau^* << \tau_G$), the disappearance of AS under 100°K would also be explained. For alloy-softened iron-nitrogen alloys these relationships have been tested [50]. Although one should expect a continuous change of the value with gradually increasing content of impurity, the experimentally found temperature dependence of the effective tension (Figs. 4a and b), and, as a result, also the sensitivity to the speed of stretching $\lambda = \partial\tau/\partial\ln(\dot{\gamma}/\dot{\gamma}_o)$ (Fig. 5), allow no differentiation according to the content of alloy.

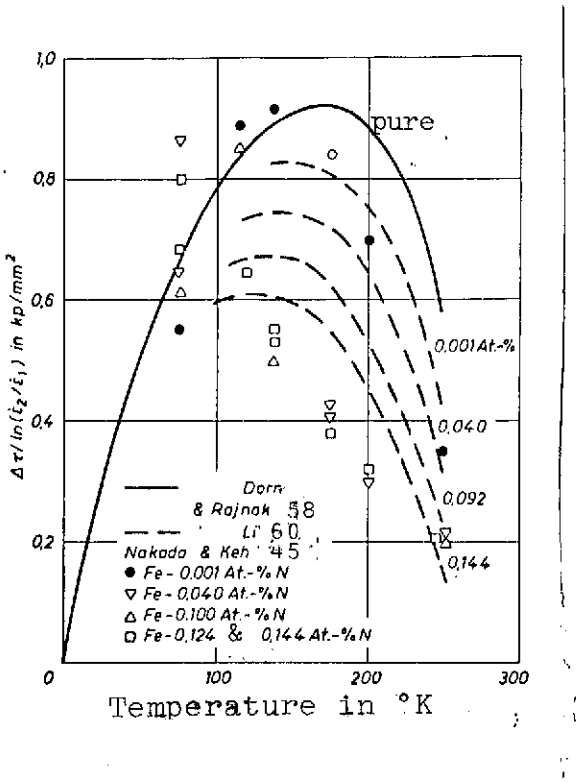

Fig. 5. The temperature dependence of the sensitivity to the speed λ in iron-nitrogen alloys [45, 50].

A theory oriented towards the indirect participation of foreign atoms involves the equilibrium configuration of dislocation slips of various lengths under externally applied tensions. The foreign atoms block the lines of dislocation; only the free slips between two such 'fail points' can overcome the Peierl's barriers and occupy a stable position in the next Peierl valley. However, if the external tension is too small to counteract the line energy of the dislocation slip, no deformation can take place. There is a given critical limiting tension τ_c (dependent on the concentration of fail points), which is independent of the temperature (the blocking at the fail

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points is taken to be independent of thermal influences). τ_c and the Peierl Tension τ_p are not additive, and, therefore, the original conditions obtained from the double kink theory will remain under a given temperature, if $\tau_p \geq \tau_c$. This is shown in Fig. 6 with broken lines. The 'pseudo-thermal' component τ_c gives the appearance of lowering the effective tension τ^*. The results of Fig. 4 are cited as evidence, although a quantitative agreement of impurity concentration and magnitude of AS in theory and experiment is not obtained.

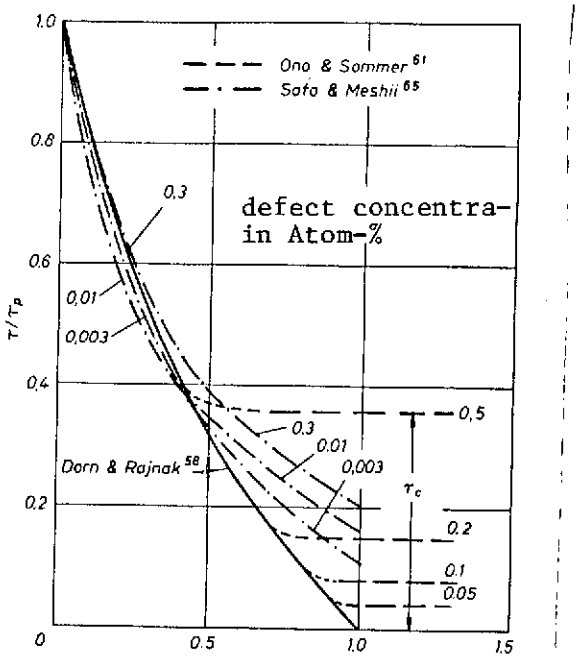

Fig. 6. The modification of the double kink mechanism by impurities, with the assumption of unchangeable anchor points (dashed lines) [61] and with mutual consideration of kink formation and overcoming the tension field of impurities (broken-point lines) [65].

The models built on the following proposition [62] are explicitly oriented to the effects of foreign atoms. If centers of tension (foreign atoms) are present in the lattice, then there

will be reactions with dislocations. These favor the formation of double kinks at the dislocations in the vicinity of the foreign atoms [5, 14, 27-29, 63, 64]. A quantitative treatment using Fleischer's principle of the reciprocal action of solution atom and dislocation has confirmed the lowering of the work of formation for kinks [23]. A new model [65] investigates kink formation as well as the lateral motion of the kink in a unified field of Peierl's- and impurity tensions. Defect centers assist the formation of kinks, as was also established elsewhere, but hinder the lateral motion of the kinks. Principally, the energy of both processes determines the overall movement of the dislocation. This apparently provides a single description of AS and mixed-crystal hardening. The result is drawn in Fig. 6 with a broken-point line: hardening takes place at high temperatures, softening /875
at low. The disappearance of AS at the lowest temperatures is based on the theory that this should only happen naturally in the vicinity of absolute zero. This model is also supported by confirming the results with iron-nitrogen and niobium-oxygen alloys. The concentration dependence of the AS is somewhat accurately met with for interstitial alloys, but not, however, for substitutional cases. There, the cluster formation of the substitution atoms is called on for explanation, and this undervalues the effective concentration, whereby they become comparable to the theoretical values.

A phenomenological proof has been sought [25] for the unity of AS and mixed-crystal hardening promoted by this theory. The many experimental data show that AS goes hand-in-hand with mixed-crystal hardening in the non-thermal region. This is shown in Fig. 7 for iron alloys: for many additives the hardening and softening are often of equal size (only for cobalt and perhaps also for chromium this relationship does not hold). The alloy atoms have a twofold effect: in the non-thermal region it is a long-range tension field extending from them; below T_0 they

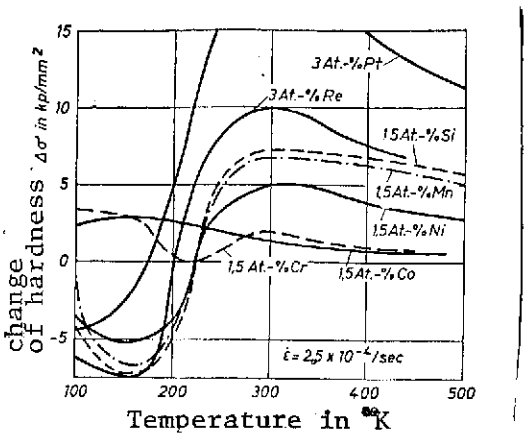

Fig. 7. AS and alloy hardening for various alloys of iron [25].

facilitate the thermal activation of dislocation mobility and bring about AS. We have sought to find correlations for AS with the help of the parameters known from the theory of mixed-crystal hardening (shear modulus defect $\varepsilon_G = (dG/dc)/G$ and atomic size defect $\varepsilon_a = (da/dc)/a$; a = lattice parameter), or the differences between shear modulus G and atomic radius R ($\Delta G = G_{basis} - G_{alloy\ metal}$ or $\Delta R = R_{basis} - R_{alloy\ metal}$). A negative ΔG would be seen as favorable for kink formation [63]. Alloy atoms larger than matrix atoms should be able to cause only small AS [34]. Both viewpoints however have been refuted by the results of many iron alloys [25] (Table I). It seems that the indications of ΔG or ΔR values are meaningless. In only one case is there a definite connection with the atomic size: if ΔR or ε_a are zero or nearly zero, there is no AS.

The increase of dislocation density. The model is dependent only on the increase in the density of free dislocations (ρ), with all quantities constant [45]. According to the Orowan equation

TABLE I. MIXED CRYSTAL PARAMETERS AND THE APPEARANCE OF AS [25, 35, 101-104]

Alloy	ΔG in 10^3 kp/mm²	ΔR in Å	ε_G	ε_a	LE in Experiment
Nb–V	−1.03	0.12	1.05	−0.074	yes
Ta	−3.3	0	1.15	0	no
Ta–Hf	1.6	−0.12	1.27 *	0.041	yes
Nb	3.3	0	−0.04 *	−0.0008	questionable
Mo	−5.2	0.07	0.49 *	−0.06	yes
W	−8.1	0.07	0.67 *	−0.046	yes
Re		0.09	0.91 *	−0.078	yes
Mo–Hf	6.8	−0.19			yes
Ta	5.2	−0.07			yes
W	−2.9	0			no
Fe–Si	4.42	−0.06	−0.11	−0.01 to −0.03	yes
Cr	−0.5	−0.01	0.32	0.02	questionable
Mo	−3.7	−0.13	0.15	0.10	yes
Re		−0.11	0.75	0.10	yes
Co	0.84	0.01	0.34	0.003	no
Ni	0.62	0.02	−1.06	0.17	yes
Cu	3.83	−0.02			yes
Au	5.65	−0.18			yes
P		−0.02	0.93	−0.014 to −0.023	yes

*Actually, $\varepsilon'_G = (1 + |\varepsilon_G|/2)$, but ε_G and ε'_G show the same tendency [25].

$$\dot{\gamma} = \rho b v \qquad (2)$$

and the empirical equation for the speed of dislocation

$$v = C\tau^{*m^*} \qquad (3)$$

τ^* must climb with falling ρ (softening!) in order that the necessary speed of stretch $\dot{\gamma}$ can remain preserved. The preexponential factor $\dot{\gamma}_0$ in the reaction equation is seen to be directly proportional to the dislocation density, and experimental measurements of $\dot{\gamma}_0$ should be a measure of the changes in ρ. The doubled cross-slips of dislocations in the neighborhood of impurity atoms and the associated proliferation of dislocations are mentioned as the cause of the increased number of dislocations [66]. /876

The model of the relationship between AS and increased dislocation density cannot be reached by exact considerations. It is based on an increase of the preexponential factor determined experimentally with interstitial alloys, for a formula derived from the reaction equation and an expression for the enthalpy of activation:

$$\ln \dot{\gamma}_0 = \ln \dot{\gamma} - T \left[\frac{\Delta\tau^*}{\Delta T} \right]_{\dot{\gamma}} \left[\frac{\ln (\dot{\gamma}_2/\dot{\gamma}_1)}{\Delta\tau^*} \right]_T \qquad (4)$$

The increase in $\Delta\tau^*/\Delta T$ (negative!) is small if there is AS. An increase in $\ln \dot{\gamma}_0$ can therefore only derive from the second term $\ln (\gamma_2/\gamma_1)/\Delta\tau^*$, the reciprocal of the speed sensitivity λ. From Fig. 5, λ is actually always smaller in alloys, and diagrams of the temperature dependence of $\ln \dot{\gamma}_0$ therefore show the increase in the region of AS [43, 45]. This empirical proof, however, proceeds from the assumption that $\Delta\tau^*$ is a function only of the

dislocation density, and does not take into account that $\Delta\tau^*$, the change in tension with a change in velocity, must also change if the mechanism of deformation in alloys is of another kind (see next section). Observations of another kind refute the view that proliferation of dislocations in alloys could be introduced through doubled cross-slips. In many cases, studies in the area of AS have found directly and clearly formed slip bands that were separated by undeformed zones, and this has often been ascribed to the result of the lack of a cross-slippage [11, 45, 67, 68]. In pure metals, in contrast, slip bands that are branched and not arranged strictly crystallographically are the rule, which point to transverse slips [27, 45, 46].

In the case of the substitution alloys iron-nickel/silicon/molybdenum, an evaluation of parameters of differential activation analysis showed that dislocation proliferation played no role in the AS [5, 69, 70].

The change of deformation mechanism. The described processes are only modifications of one definite mechanism characteristic of the b. c. c. lattice. For it, the double kink mechanism, J. E. Dorn gives us [58, 59]

$$\tau^* = \tau_p[1 - (T/T_0)^{1/2}] \qquad (5)$$

J. E. Dorn demonstrated in his very first work [58] on the subject that his model does not adequately describe the process in the region of T_0 (it provides too steep a temperature dependence), and suggested later that B. Escaigs' recombination mechanism [56] could define the gradual transition to non-thermal behavior.* This model would have no meaning in pure b. c. c. metals, since because of the high breakdown energy, cleavage is negligible.

*We have already mentioned earlier another explanation, the model of internal tensions.

However, in alloys it could prove correct. The recombination model has been tested or considered in the cases of the results from iron-manganese [59], iron-silicon [17], and niobium-tungsten [30] alloys, whose tendency to AS has been established. Discontinuities in the relations $\Delta H - T$ or $\Delta H - \tau^*$ show that at about 200°K the return to the double kink mechanism is probable.

A variation in the deformation mechanism asserts itself in principle in the display of the temperature dependence of λ. Each discontinuity demonstrates that the straightforward conditions set forth for a single mechanism by the reaction equation is disturbed. If two mechanisms are involved in separate temperature ranges, stages occur in $\lambda(T)$, because different temperature dependences exist for varying mechanisms, and moreover their activation enthalpies can be different. If the λ-values of two materials are different at a given temperature, then it follows from the constancy of $\dot{\gamma}_o$ that the material with the smaller λ has a smaller dependence of tension on temperature. AS therefore comes into play if λ of the alloy is small compared with the value for the pure metal. In actuality, it is not always easy to arrive at this orderly relationship, since frequency factors are not known for all mechanisms. However, a discontinuity in the $\lambda(T)$-curve is not an infallible indication of a change in mechanism. For example, it is very marked in the curves for iron-nickel/silicon alloys in similar fashion to Fig. 5, yet there the basis for AS is rather the easing of double kink formation. In a true change of mechanism, it is to be expected that the $\Delta H - \tau^*$ relation is not just numerically shifted as for iron-nickel/silicon [5, 21], but is fundamentally of another type. Only in combination with the discontinuity in λ should this be an unquestionable indication of the change.

We find both of these conditions in vanadium-titanium alloys, and it has been shown that the transitions in $\lambda - T$ (Fig. 8) and in $\Delta H - \tau^*$ (Fig. 9) diagrams stand in a causative relationship.

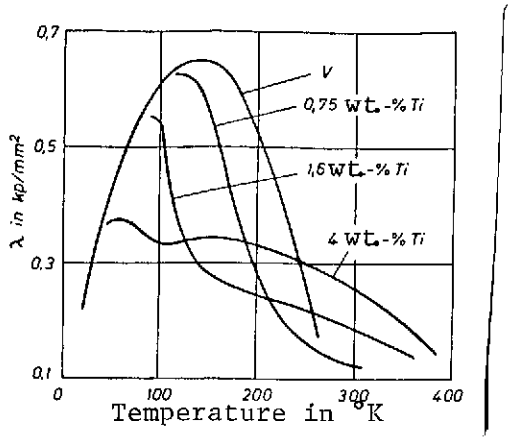

Fig. 8. The temperature dependence of the speed sensitivity in vanadium-titanium alloys [36].

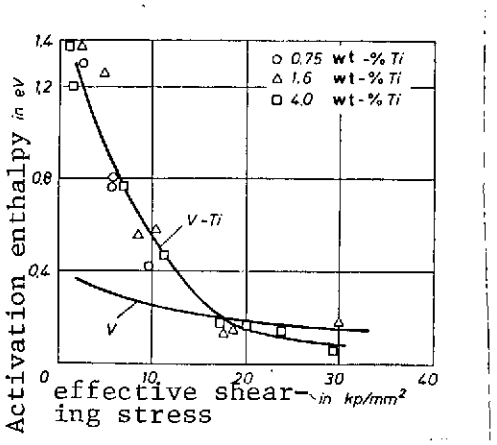

Fig. 9. The tension dependence of the activation enthalpy of vanadium-titanium alloys [36].

Between 100 and 300°K (at low tensions) R. L. Fleischer's $c^{1/2}$ rule [71] is useful, and the activation enthalpies of the alloys show a course sharply divergent from the double kink mechanism of pure vanadium. Below 100°K the ΔH-values of the alloys again correspond with those of the pure vanadium and they are characteristic of the double kink mechanism. In spite of the presence of both of these processes of deformation, the AS in the

system vanadium-titanium is not immediately perceptible. The impurity mechanism determines the position of T_o, and, limited by its higher activation enthalpy, T_o is displaced to higher temperatures. Therefore, the λ of the alloys is smaller than λ of pure vanadium only in a limited temperature range, and since T_o of the alloys lies higher, λ of the alloys is still measurable when λ of vanadium has already become zero (Fig. 8). In the temperature range between the two T_o's, the τ^* curve of the alloys is already climbing, and therefore no AS is found also if $\lambda_{V-Ti} < \lambda_V$ (Fig. 10). The AS between 30 and 100°K for the 4 wt.-% alloy is explained by the influence of kink formation [36].

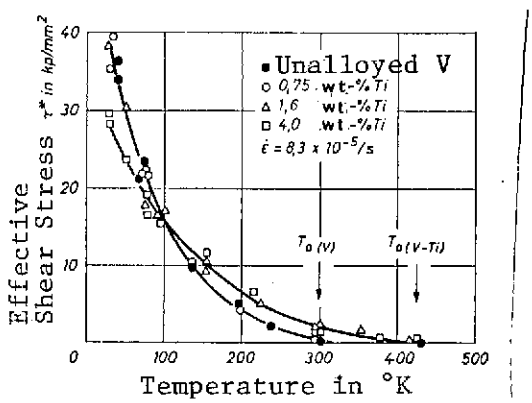

Fig. 10. The temperature dependence of the effective shear stress of polycrystalline vanadium-titanium alloys [36].

In interstitial alloys, if the interstitially dissolved elements are viewed as determining deformation, the low temperature behavior is to be described by R. L. Fleischer's mechanism of tetragonal deformations [71]:

$$\tau^* = \tau_o^*[1 - (T/T_o)^{1/2}]^2 \qquad (6)$$

(τ_o^* is the effective tension at 0°K). The two relationships (5) /877 and (6) give different curves: Fleischer's theory gives the smaller temperature dependence $\partial\tau^*/\partial T$ in the vicinity of T_o

(Fig. 11). The equations describe fairly well the experimental

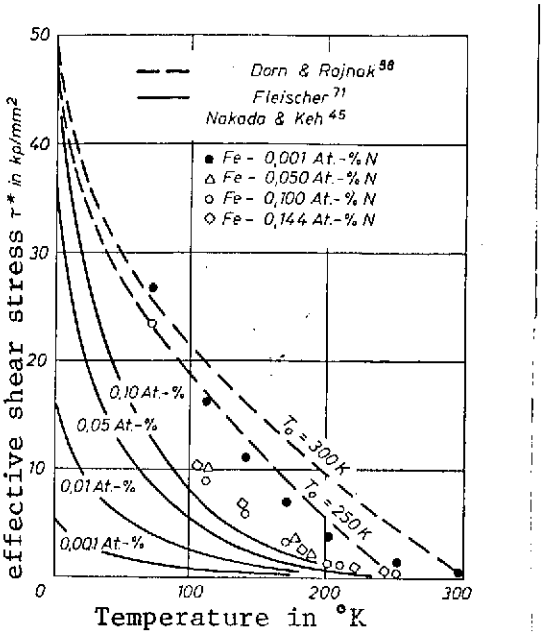

Fig. 11. Experimental temperature dependence of the shear stress in comparison with theoretical predictions [50].

temperature dependence of the shear stress of ultrapure iron and iron enriched with 0.1 Atom-% N. The fact that the softening effect disappears below 100 to 150°K is in agreement with the observation that the theories of reciprocal action of dislocations and impurities are no longer applicable at low temperatures [50]. The low-percentage nitrogen alloys cannot be described with R. L. Fleischer's mechanism [71], for, experimentally, there is no significant concentration dependence of AS (see also Figs. 4a and 4b). It has been attempted to explain this by a saturation by impurity atoms in the region of dislocation [50]. This idea of the

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*The theoretical τ^* - T curve for the double kink mechanism is still to be corrected according to J. C. M. Li's theory [60] of the internal tensions, whereby τ^* drops.

alternation of deformation mechanism in interstitial alloys also runs up against other difficulties. Indeed, as we have seen, Fleischer's theory and also the further development of Frank [72] agree with the experimental results between 100 and 300°K by assuming a formal saturation (Figs. 5 and 10) [50, 73], yet this stands opposed to the frequently represented view that several measurements (activation surfaces [74], microexpansions [75]) are not to be brought into harmony with the actual small concentrations.

AS also occurs if incompatible excretions are distributed through the lattice [13, 18, 19]. The situations are largely unexplained. For example, in an addition of substitutionally soluble elements in excess, the softening should depend on the kind and structure of the excretions [19]. There is also AS in a matrix with dispersed foreign phases (for example, molybdenum-Al_2O_3/MgO) [48] and in alloys with interstitially soluble elements if these exceed the limits of solubility [47, 50]. The well-known models certainly will no longer serve here, yet the agreement of reduced effective stress and the activation parameters is again valid according to equation (2): the activation volumes and enthalpies are too high [18, 19, 23, 24] to be consistent with the double kink or impurity mechanisms. In the case of molybdenum alloys--which have been measured at 2 to 4 eV--diffusion-limited reactions such as the non-conserving progression of Jog that have arisen at inclusions are determining factors for the deformations [48, 76].

The 'Scavenging' Model. The 'clean sweep' of the metal /878
matrix, performed by the alloy additives, is the earliest explanation for AS. Ever since 1944, this idea was called upon for an explanation of the phenomena in substitutional iron-based alloys [8, 11, 77], and later for chromium- and molybdenum-(platinum metal) [78, 79], tungsten-rhenium, and niobium substitution alloys [80]. Later the argument was also expanded to interstitial

solutions [47]. Thus, it was postulated that in niobium-nitrogen alloys, the addition of oxygen can lead to complex formation with other, already present, impurities. Cluster formation between oxygen and carbon/nitrogen has also been confirmed in factual experiments [81]. As proof of the scavenging theory the fact was brought forth that the AS effect is never seen in progressive purification of a metal by zone melting and high vacuum incandescence. AS may be always observed then, only if the alloying constituent is mixed inversely with the 'pure' metal. The impurities (carbon, nitrogen, etc.) in this model are taken to be of the short-range tension-determining barrier type. If they are removed from the lattice by their chemical affinity to the alloy element, they are no longer available as a solution-hardening constituent, but can now act more indirectly as excretions on the non-thermal components. These views have been criticized; see [82]. Quenching studies on iron-carbon alloys seem also to be strong counterevidence: quenched material (the growth in dissolved carbon was determined with a torsion pendulum) had the smaller low-temperature hardness and the smaller slope $\partial\tau^*/\partial T$ [21, 83]. In mechanical studies of vanadium-titanium alloys, which also were combined with the determination of Snoeks' friction maxima, it was found that no AS is brought forth by initial scavenging. It only appears in higher-percentage solutions if scavenging ended long previously [36, 84]. Even the interpretation of the scavenging model as an irreversible process seems not unequivocal, since, in at least two experiments, the yield points of iron alloy--softened with nitrogen or carbon additives--rose again after an incandescence purification in hydrogen [46, 85].

The latest information justifies the assumption of the scavenging process in spite of everything. In tantalum-rhenium [32] and niobium-molybdenum alloys [4, 34, 86], of which the former are considered to be 'classic' examples of AS, it is completely lacking if a mixed crystal completely purified of

interstitial elements is studied. AS also is lacking if an unpurified alloy is cooled quickly [32]. It is likely in that case that there is not enough time for the reciprocal action and complex formation to take place.

The described contradictory aspects have not yet been treated coherently in the literature.

The original idea of scavenging depended on the reciprocal effect of elements with high chemical affinity for one another. However, molybdenum is not viewed as an oxide-, carbide-, or nitride-forming if it is dissolved in niobium. The motive force for scavenging here must be not chemical but of a geometric nature, i.e., the potential fields of the interstitial and substitutional atoms combine to diminish the overall potential energy [34]. This kind of lattice purification has been called 'pseudo-scavenging' [87]. Reciprocal effects of this kind can be derived from measurements of the internal friction. At today's level of knowledge, there are such reciprocal effects in the majority of cases [87], yet the correlation of atomic reciprocal action and mechanical AS effect is not at all as conclusive as for example in the case of the tantalum-rhenium-nitrogen alloys [88]. The insensitivity of the softening to changes in the carbon content in iron-silicon and iron-nickel alloys [21] is not reconcilable with the statement about the internal friction.

Very recently the role of complex formation has also been examined, in interstitial alloy systems. What at first had been only postulated was confirmed by experiment: there is required, in fact, a residual amount of a second interstitial impurity before a systematic addition of oxygen or hydrogen will cause AS (Fig. 12) [89-91].

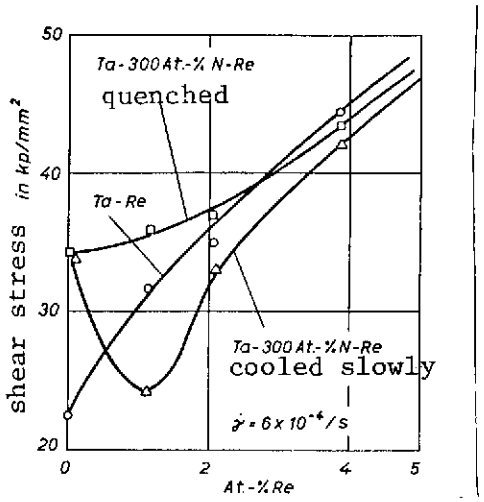

Fig. 12. The influence of purity and heat treatment on AS at 77°K in tantalum-rhenium single crystals [32].

For a long time it was not understood why AS was only found in a few interstitial alloys. Systematic investigations of tantalum-nitrogen [92-94], tantalum-oxygen [92], niobium-nitrogen [95], niobium-oxygen [96], niobium-hydrogen [97], and vanadium-oxygen [98], brought no indication of AS. This is understandable in the light of the latest work with combined interstitial impurities, since in all these studies the starting material was of the highest purity.

The influence of the electron concentration. Early interpretations of AS settled on the increased number of valence electrons in alloys and the reduced solubility of the metalloid [10, 78]. This corresponds in a certain sense to the scavenging idea: the result is a lattice free of impurities. This explanation of low temperature hardness depends on extrinsic factors.

However, it has been inferred also from the relation between electron configuration and the magnitude of the Peierl Tension

[14]. Alkali metals, which have only s-electrons in their outer shells, show a weak increase in the yield point at low temperatures. The unfilled and non-spherical d-shells in transition metals should exhibit high Peierl forces, and every filling-up with d-electrons should lower the Peierl Tension. In alloys that have a larger number of s- and d-electrons than the basis metal, AS should therefore predominate [41]. Following this consideration, for example, rhenium, osmium, iridium, or platinum in molybdenum cause AS, while hafnium and tantalum harden molybdenum. There are also definite c/a concentrations which should display the border between softening and hardening. This statement cannot be adopted uncritically: in the consideration of the research data, the totality of the tension ($\tau_G = \tau^*$) was used and it was ignored that the temperature dependence $\partial\tau^*/\partial T$--from the earlier expositions itself an important determinant of AS--also is reduced in molybdenum alloyed with hafnium and tantalum.

A further interpretation of the influence of the electron /879
configuration stems from the partially covalent bonding character in metals with unfilled d-shells [20]. In analogy to materials with covalent bonds, it is assumed that slip planes can be activated only at high temperatures. At lower temperatures there comes about the cooperation of definite slip systems with higher critical shear stresses. In a crystalline aggregate, where the compatibility conditions at the grain walls are more difficult to set up, the flow potential is increased. In alloys with suitable elements, which can fill the outer d-shells, the covalent bonding portion diminishes, more slip systems are active, and the measured flow potential shows the AS effect. This model seems to depend too much on the conditions in the polycrystal to have general applicability. It also ignores the fact that the addition of elements which dilute the concentration in the d-shells can bring forth AS.

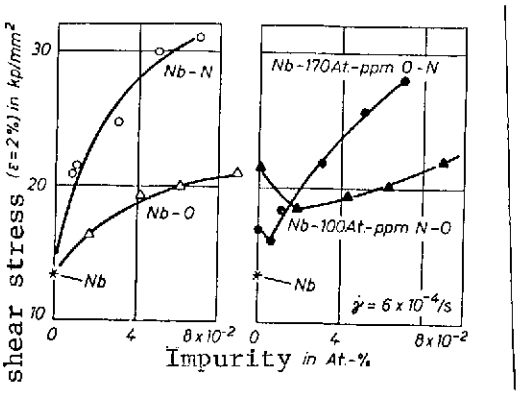

Fig. 13. The influence of purity on the AS at 113°K in single crystal niobium alloys [91].

Closing Considerations

The theories for explaining AS rest on:

(1) The influence of the Peierl Tension or of the double kink mechanism through alloy atoms;
(2) The increase in dislocation density in alloys;
(3) Alternation in the deformation mechanism;
(4) The purification of the lattice through chemical or geometric reciprocal action between alloy atoms and dissolved impurity atoms;
(5) The change in the number and configuration of free electrons.

Theories (2) and (5) probably deserve the least consideration. The foundations rest on a series of assumptions that do not hold up with the results of other studies. Thus, for example, the increased doubled transverse slips and the dislocation proliferation are not proved, and the interpretation of the frequency factor is not unconditionally correct. The electron concentration, the filling of the s- and d-shells and the resulting changed

solubility for interstitial elements do not really constitute a mechanism, but only the hypotheses for one. They have been tested in certain cases, but cannot be generalized. AS is more the rule than the exception in low temperature behavior, and the relative place of the basic and additive metals in the periodic system is unimportant if all cases of AS are taken into consideration.

Point (1) makes use of both a direct and an indirect reduction of the Peierl Tension. The first possibility, the changing of τ_p through the modulus, is not likely, since the necessary irregular changing of the modulus would have to be of a similar dimension as the irregular changes in the shear stress. There remains the indirect reduction of the tension through the influence of the alloy on the formation of kinks. Doubtless there is a connection with mixed crystal hardening: the degree of the thermally activated AS depends on how far an alloy element is able to strengthen the non-thermal tension component of the base metal. In contrast to statements in the literature [25, 63], one can obtain valid statements for all cases, either from the difference of the elastic constants, from the atomic diameter, or from the modulus defect, because positive as well as negative values are compatible with AS (Table I). That the atomic size defect is of influence seems to be certain in only one special case; namely, when $\varepsilon_a = 0$, there is neither mixed crystal hardening nor AS.

The alternation in deformation mechanism mentioned in point (3) as a cause for AS seems to be documented fairly often. The low temperature deformation of pure b. c. c. metals by the double kink mechanism with its steep $\tau^* - T$ dependence is described frequently. Theoretically, smaller temperature dependences are observed based on an impurity mechanism, if interstitial

impurities are alloyed. For many substitution crystals the presence of a second mechanism in the region of AS is demonstrated through increased activation enthalpy. An intrinsic characteristic of AS--its disappearance below 100°K--is in agreement with the empirically found return to the double kink mechanism.

Indications for the scavenging model described in Point (4) are found in substitutional and interstitial alloys. If the theories (3) and (4) are not considered together here, this fits in with the way scavenging has been interpreted in the literature. One does not proceed from the double kink mechanism, but assumes that the low temperature deformation is determined by the interstitially dissolved foreign atoms. Scavenging reduces the number of these lattice impurities. The shear stress is reduced with progressive purification, assuming that the deformation mechanism itself is not changed thereby. The theory seems to be confirmed by the experimentally found complex formation, however, our understanding fails over the kind of dislocation progagation in the purified and in the initial condition. If we consider the reciprocal effects of different kinds of atoms, we should also look at the behavior of these reaction products in deformation. It could be that the basis for AS is not the lattice purification, but the dislocation progagation influenced by the excretions. Therefore, in the strict sense, we should not speak of scavenging as a 'mechanism,' but it provides the suppositions for the course of the deformations according to a specific mechanism, in a basic way similar to that of the electron concentration.

There should still be pointed out an unclear point in connection with AS in iron-nitrogen alloys. The solubility for carbon and nitrogen is known to be extremely small at room temperature, so that in the temperature range of AS even with medium contents, excretions are possible. Since the measured activation enthalpies of deformation for these iron alloys are not abnormally high, but

are similar to the theoretical values for the Fleischer mechanism, the conclusion suggests itself that the excretions must be of a coherent nature and can be traversed [72, 100]. The presence of coherent particles has been confirmed in iron, and in iron alloys [101]. In this way, it could also be explained that the size of the AS in iron-nitrogen systems does not change continuously with the nitrogen content [45]. The presence of coherent excretions has also been set forth as an explanation for the discrepancies between theory and experiment for mixed crystal hardening [101]. In a continuation of an earlier thought, this could also be responsible for the fact that no correlation is possible between AS and the different nuclear sizes of the crystal.

In an earlier paragraph, the question was posed whether AS could be declared to be different in substitutional and interstitial alloys, or whether it is a question of different groups of substitutional alloys. The first question must be denied. One experimental certainty, the disappearance of AS below 100°K, can be found in interstitial and substitutional alloys. Therewith is lost one important argument for separating into two groups. If,also,the details of the model presentation can be altered according to the type of mixed crystal, then an overall generalization on the groundwork mentioned earlier seems possible. Scavenging or pseudo-scavenging of the matrix could initiate a change of the mechanism of deformation (details of the dislocation propagation would still have to be clarified), and, in fact, this model fits both types of mixed crystals. However, apparently this mechanism cannot be carried over directly to all substitutional alloy systems. Frequently there is no evidence for reciprocal effects of chemical or geometric nature between foreign atoms and matrix atoms. In these cases, the dominance of the earlier-mentioned direct influence of the Peierl Tension on the shear stress (with all its limitations) or the easing of kink formation, are to be considered.

The Director of the Institute, Herr Professor Dr. H.-P. Stuwe, is heartily thanked for his help. He has promoted the accomplishment of the work with criticism and advice.

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